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Mass-polarization effects in the $1s2s\ ^1S$ and 3S states of helium

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From a laser spectroscopic study of isotope shifts in the $1s2s\ ^1S, ^3S \rightarrow 1snp$ transitions in ^3He and ^4He and of the hyperfine structure in the $1snp$ Rydberg states of ^3He we have derived highly accurate information on mass-polarization effects in the $1s2s\ ^1S$ and 3S states. The experimental results, which are in good agreement with recent, high-precision variational calculations, confirm an isotope dependence of $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$.

Spectroscopic studies of two-electron atoms give more and more insight in the effects of correlations in the motion of both valence electrons.^{1,2} It is well established that electron correlations play an important role in the doubly-excited states of helium and in the ground and low-lying states of alkaline-earth atoms and several models have been developed to treat their influence.³⁻⁶ One measure of the degree of correlation is the inner product of the linear momenta $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$ of both electrons. Its measurement provides an important, independent test of model calculations.

The mass-polarization shift is defined as $\epsilon_M = (m_e/M) \langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$, with m_e the electron mass and M the nuclear mass. This mass-polarization shift is directly related to the so-called specific mass shift (SMS), which gives a substantial contribution to the isotope shift in atomic-level energies. When comparing energies of different isotopes of the same element, shifts arise from differences in nuclear mass and volume, slightly affecting the electronic orbitals. The volume shift is due to the fact that an electron, especially in an orbital with low angular momentum l , experiences different charge distributions close to the nucleus. For light elements, such as helium, the volume effect may be neglected. The mass shift is composed of the normal mass shift (NMS) and the specific mass shift (SMS). NMS accounts for differences in reduced mass and can be calculated exactly. It follows that in helium a determination of level-energy shifts directly yields values for SMS.

The quantity which can be investigated experimentally, however, is the transition isotope shift (TIS), i.e., the difference in energy shifts of the levels involved in the transition. The deduction of level shifts from measured isotope shifts in transitions between low-lying states is a complex problem.⁷ When the upper state involved in the transition is a highly excited Rydberg state, the problem

simplifies considerably. For helium Rydberg states $1snl$, it is easily shown that the inner product $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$ scales as n^{-3} (approximately), and consequently SMS converges to zero with increasing value of the principal quantum number n (see, e.g., Ref. 1).

In this Rapid Communication we report values for SMS and mass-polarization effects of the $1s2s\ ^1S$ and 3S states of helium as deduced from a high-resolution laser spectroscopic study of TIS in the transitions $1s2s\ ^1S, ^3S \rightarrow 1snp$ for ^3He and ^4He . In the experimental setup an atomic beam of an isotopically enriched mixture of ^3He and ^4He was used. The gas mixture was expanded from a quartz tube nozzle via a skimmer into the excitation chamber. To populate the metastable $1s2s\ ^1S$ and 3S states a low-current discharge was maintained between a tantalum needle inside the quartz tube and the skimmer. In the excitation chamber $1snp$ Rydberg states were excited from these states with a laser beam, perpendicularly intersecting the atomic beam. To eliminate stray electric fields the excitation region was carefully shielded with a metal box. The uv laser radiation required for the excitation was produced by intracavity frequency-doubling of a spectra physics 380D ring dye laser (linewidth < 1 MHz). For the excitation from the $1s2s\ ^1S$ state, 5 mW of uv light in the wavelength region around 313 nm was generated using the dye Rhodamine B and a temperature-tuned nonlinear rubidium dyhydrogen phosphate (RDP) crystal, whereas for the excitation from the $1s2s\ ^3S$ state, 3 mW in the region 260–263 nm was available from a Coumarine 6 laser frequency doubled in potassium dyhydrogen phosphate (KDP). He ions produced from Rydberg states by field ionization were focused into a quadrupole mass filter and detected by an electron multiplier. Calibration of wavelength scans was performed with a Fabry-Perot étalon, length stabilized with an I_2 -stabilized He-Ne laser. Its free spectral range was 148.9553(6)

MHz, accurately calibrated on the separation of the Na D_1 and D_2 lines. More details of the setup can be found elsewhere.^{8,9}

In Fig. 1, as an example, the excitation $1s2s\ ^1S \rightarrow 1s40p$ is shown. The experimental linewidth of 15 MHz full width at half maximum (FWHM) was entirely due to residual Doppler broadening. This spectrum of 60 GHz width shows the transition to 1P_1 in ^4He (single peak at the high-frequency side) as well as two hyperfine transitions of ^3He (nuclear spin $I = \frac{1}{2}$). The peak at the low-frequency side contains two hyperfine components with primarily 3P character (with hyperfine quantum numbers $F = \frac{1}{2}, \frac{3}{2}$) excited because of hyperfine-induced singlet-triplet mixing. The remaining peak is the regular 1P_1 excitation, also with unresolved $F = \frac{1}{2}, \frac{3}{2}$ components. Since the hyperfine interaction of the np electron varies approximately as n^{-3} individual hyperfine components could not be observed for high n . Although the $1s2s\ ^1S$ state is populated more efficiently than the $1s2s\ ^3S$ state in our discharge,⁹ we did not observe singlet Rydberg states with $n < 30$ as a consequence of the short radiative lifetime of these states and our detection of He ions produced by field ionization. Field ionization was induced a few mm downstream from the laser-atomic-beam interaction region, so low- n Rydberg atoms with a large 1P fraction in their wave function will decay to the $1s^2\ ^1S$ ground-state hampering detection.

Hyperfine structure of ^3He is a complicating factor in the determination of TIS. It was studied in detail by Vassen and Hogervorst,⁸ who were able to explain their observations, including the effects of hyperfine-induced n mixing, using a three-channel, two-limit (hyperfine energies of the $^3\text{He}^+$ ground state) quantum-defect analysis. Following the same analysis the center of gravity of the ^3He hyperfine multiplets was obtained resulting in the determination of the isotope shift in transitions from $1s2s\ ^1S$ and 3S states. As outlined before, the specific mass shift in the transition may be deduced from TIS by subtracting NMS, neglecting volume effects. To calculate NMS we used $m_e = 5.485803 \times 10^{-4}$ amu, M_4

TABLE I. Experimental values (in MHz) of the specific mass shifts of $^3,^4\text{He}$ in $1s2s\ ^3S \rightarrow 1snp\ ^3P_2$ and $1s2s\ ^1S \rightarrow 1snp\ ^1P_1$ transitions deduced from the transition isotope shift by subtracting the normal mass shift. The error is one standard deviation. Values for triplet series were reported earlier by Vassen and Hogervorst (Ref. 8).

n	Transition SMS (expt.) $1s2s\ ^1S \rightarrow 1snp\ ^1P_1$	Transition SMS (expt.) $1s2s\ ^3S \rightarrow 1snp\ ^3P_2$
20		2206.1(30)
30		2200.3(30)
40	2790.5(40)	2193.9(30)
45	2789.7(40)	2192.5(30)
51	2793.0(40)	2188.5(30)
52	2795.3(40)	
56	2791.8(40)	
60		2189.4(30)

$= 4.001506079$ amu, and $M_3 = 3.014932149$ amu.¹⁰ Experimental values of SMS for different values of n for transitions from $1s2s\ ^1S$ as well as 3S are given in Table I. From this table it can be seen that the specific mass shift converges for high n to a constant value, the SMS of the metastable state.

More accurate isotope shift measurements around $n=50$ are feasible as was already pointed out by Vassen and Hogervorst.⁸ As a consequence of the large value of TIS the $1snp$ peak of ^4He nearly coincides with the $1s(n+1)p$ peaks of ^3He in the interval $n=50-55$. An example is shown in Fig. 2 for the $1s2s\ ^1S \rightarrow 1s51p\ ^1P_1$ (^4He), $1s52p$ (^3He) transitions. In this case the small frequency separations may be measured with higher accuracy. The energy difference between adjacent n levels can in principle be calculated precisely (see, e.g., Ref. 11) resulting in highly accurate values for TIS. We leave this for future studies.

Several high-precision variational calculations for the mass-polarization shifts in low-lying states of helium are available.¹²⁻¹⁴ Kono and Hattori¹³ give values for $1sns$

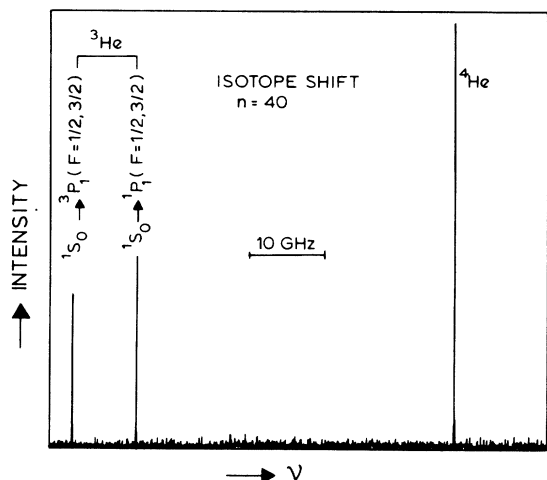


FIG. 1. Isotope shift between ^4He and ^3He in the $1s2s\ ^1S \rightarrow 1s40p$ transition at 312.9 nm.

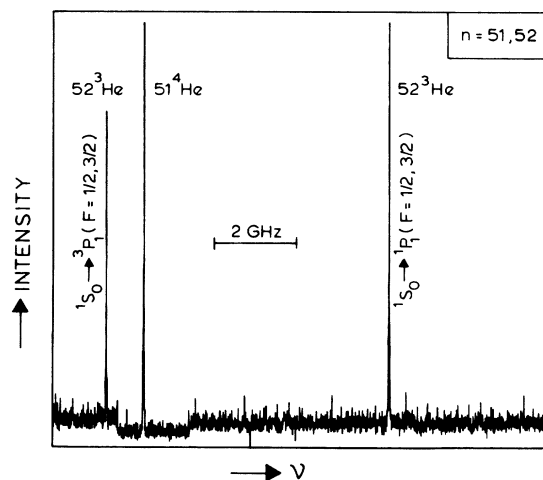


FIG. 2. Spectrum $1s2s\ ^1S \rightarrow 1snp$ for $n=51$ in ^4He and $n=52$ in ^3He at 312.6 nm.

TABLE II. Experimental and calculated values (in MHz) of the SMS ($=\epsilon_3 - \epsilon_4$) of the $1s2s\ ^1S$ and 3S states of helium.

	SMS (expt.)	SMS (calc.) ^a	SMS (calc.) ^b	SMS (calc.) ^c
$1s2s\ ^1S$	2793.0(40)	2804.9(1)	2790.75(1)	2789.87(1)
$1s2s\ ^3S$	2189.6(25)	2196.400(1)	2189.89(1)	2188.68(1)

^aReference 13.^bReference 14.^cReference 14 with relativistic corrections of Refs. 15 and 16.

$1s^3S$ and $1snp\ ^1,^3P$ series up to $n=8$. Assuming no mass dependence of $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$, the SMS for these levels may be calculated directly from $\text{SMS} = \epsilon_4(M_4 - M_3)/M_3$.

From isotope shift calculations for the $1snp$ levels in between $n=3$ and 8 we calculated the contribution of the $1snp$ levels with $n > 8$ to the specific mass shift of the transition using the extended scaling law $a(n)^{-3} + b(n)^{-5} + c(n)^{-7}$ (Ref. 8) where higher-order odd powers are neglected. It follows that the contribution of the $1snp$ level in the triplet case is, e.g., -2.0 MHz for $n=40$, -1.0 MHz for $n=51$, and only -0.5 MHz for $n=60$, whereas in the singlet case it is $+1.9$ MHz for $n=40$ and $+0.7$ MHz for $n=56$. Correcting the transition SMS of Table I with these $1snp$ contributions, the SMS of the $1s2s\ ^1S$ and 3S states are obtained. Averaged

values are given in Table II. It is important to note that the SMS of these states is the difference in the mass-polarization shifts $\epsilon_3 - \epsilon_4$.

From the calculations of Kono and Hattori,¹³ assuming again no mass dependence of $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$, the SMS of the $1s2s\ ^1S$ and 3S states may be evaluated. These values are included in Table II. A systematic difference between experiment and these calculations is obvious. The calculated value for $1s2s\ ^3S$ state is 6.8 (2.5) MHz higher than the experimental value, whereas for the $1s2s\ ^1S$ the difference is 11.9 (4.0) MHz. Recent theoretical calculations of Drake¹⁴ for the $1s^2$ and $1s2s$ levels in H^- and He using the mass-polarization operator $(\mu/M) \mathbf{p}_1 \cdot \mathbf{p}_2$ [μ is the reduced electron mass $(m_e M)/(m_e + M)$], however, show a small mass dependence of $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$:

$$\epsilon_M(1s2s\ ^3S) = 0.007442130706(\mu/M) - 0.0574958(\mu/M)^2,$$

$$\epsilon_M(1s2s\ ^1S) = 0.009503864419(\mu/M) - 0.135277(\mu/M)^2,$$

in units of $2R_M$. Here $R_M = (1 - \mu/M)R_\infty$, with R_∞ the Rydberg constant. [Note: in Ref. 8 the relation for $\epsilon_M(1s2s\ ^3S)$ of ^3He is not correctly transformed into units of cm^{-1} as R_M for ^4He was substituted. This only slightly affects the calculated value in that work.] From these relations ϵ_4 and ϵ_3 are calculated. The values of $\epsilon_3 - \epsilon_4$ (equal to SMS) for the $1s2s\ ^1S$ and 3S states are also shown in Table II. They are in agreement with experimental values, thus confirming the mass dependence of the inner product $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$. This mass dependence is negligible for the SMS of highly excited $1snp$ states. Finally, relativistic corrections have to be taken into account. Drake and co-worker^{15,16} calculated the correction for the SMS of the $1s2s\ ^3S$ state to be -1.21 MHz. Following the same procedure we find a value of -0.88 MHz for the

relativistic correction for SMS of the $1s2s\ ^1S$ state. In the last column of Table II the relativistically corrected values of SMS of $1s2s\ ^1S$ and 3S states are given.

The conclusion is that the measured differences in mass-polarization shifts of the isotopes ^3He and ^4He both for the $1s2s\ ^1S$ and 3S states are in good agreement with the recent calculations of Drake. A mass dependence of the inner product $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$ of the momenta of the two electrons is confirmed. The present results support the validity of these variational calculations for two-electron atoms.

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